

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of: **Takeshi MINAMI et al.**

Art Unit: **1793**

Application Number: **10/583,152**

Examiner: **Diana J. Liao**

Filed: **June 16, 2006**

Confirmation Number: **2564**

For: **CATALYST FOR MANUFACTURING SYNTHESIS GAS AND METHOD OF
MANUFACTURING SYNTHESIS GAS USING THE SAME**

Attorney Docket Number: **062610**

Customer Number: **38834**

SUBMISSION OF DECLARATION UNDER 37 C.F.R. § 1.132

Mail Stop: Amendment
Commissioner for Patents
P. O. Box 1450
Alexandria, VA 22313-1450

June 22, 2009

Sir:

This paper includes an attached declaration under 37 C.F.R. § 1.132 which provides additional evidentiary support to the remarks provided in the Amendment filed March 9, 2009.

If this paper is not timely filed, Applicants respectfully petition for an appropriate extension of time. The fees for such an extension or any other fees that may be due with respect to this paper may be charged to Deposit Account No. 50-2866.

Respectfully submitted,

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Attachment: Declaration under 37 C.F.R. § 1.132

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DECLARATION UNDER 37 C.F.R. §1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Takeshi Minami, a citizen of Japan, hereby declare and state the following:

1. Since 1984, I have been employed by CHIYODA CORPORATION and assigned to the research center of the company. In the research center, I have been involved in the developments of:

a process for butanol production by fermentation for the use as fuel additive (-1987);

a technology for effective utilization of beer malt residue (1989-91) including a pilot plant test (1991);

a catalyst and a process for plant oil modification (liquefaction of solid oil) using an immobilized enzyme catalyst (1991-92);

a catalyst for acetic acid synthesis using an immobilized complex catalyst (1993-) including a pilot plant test (2000) and a first commercial plant design (2004);

a catalyst and a process for dimethyl carbonate synthesis using a complex catalyst (1994-97);

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a technology for decomposition of dioxin at ambient temperature (1999-2000);

an improved process for acetic acid synthesis (2001-);

a novel synthesis gas production process with regeneration of CO₂ reforming catalyst (2001-02); and

a catalyst and a reactor for synthesis gas production by D-CPOX process (2002-).

2. I have read and am familiar with the above-identified patent application as well as the Official Action dated January 26, 2009 in the application.

3. I have read and am familiar with the contents of cited references, U. S. Patent Nos. 5,898,014 to Wu, and 6,376,423 to Yagi; and U.S. Published Application No. 2002/0115730 to Allison, cited in the Official Action in the above-identified application.

4. Under my supervision and control, I conducted experiments to obtain data for demonstrating that a TWC catalyst, generally equivalent to the catalyst of Wu, does not exhibit such high selectivities of at least 90% for both of CO and H₂.

5. The catalyst support (carrier) prepared and used in the examples of Wu is made from a co-precipitate of zirconia and rare earth elements (ceria, praseodymia and neodymia). Accordingly, a co-precipitate of ceria and zirconia (manufactured by Daiichi Kigenso Kagaku Kogyo CO., LTD.) was baked together with magnesia to prepare a catalyst support (catalyst support A) and used in the performance test.

Further, in order to confirm that the above material of Daiichi Kigenso Kagaku Kogyo CO., LTD. is a co-precipitate of ceria and zirconia, a catalyst support (catalyst support B) was

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prepared by baking a co-precipitate of ceria and zirconia together with magnesia and X-ray spectra of support A and support B were compared with each other.

Preparation of catalyst support A

1. Twenty-five grams of a co-precipitate of ceria and zirconia manufactured by Daiichi Kigenso Kogyo CO., LTD. (in Osaka, Japan). 21.02g of $\text{Mg}(\text{OH})_2$ and 2.37g of graphite carbon were mixed and compression molded at 150kN for 5 minutes to form a molded product with a size of 25mm ϕ .
2. The molded product was baked in air for 6 hours at 1200°C with a baking rate of 100°C/hr.
3. The baked, molded product (catalyst support A) had a second/first ingredient ratio of 0.332 mol/mol, and a third/first ingredient ratio of 0.165 mol/mol.

Preparation of catalyst support B

1. Into 2 liters of water were dissolved with stirring 126g of $\text{Ce}(\text{NO}_3)_6 \cdot 6\text{H}_2\text{O}$ and 108g of $\text{ZrO}(\text{NO}_3)_2 \cdot 2\text{H}_2\text{O}$.
2. Into the above-formed solution was added slowly with stirring aqueous ammonia (28% by weight) until the pH value of the solution becomes 8.5 to cause co-precipitation of the solutes to occur. The rate of stirring was 250 to 270 rpm.
3. The solution having caused co-precipitation was filtered by means of an aspirator.

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4. The obtained co-precipitate was put into 2 liters of water, then stirred at a rate of 250rpm for 5 minutes, and filtered. This operation was repeated three times to wash the co-precipitate.

5. The washed precipitate was dried at 60°C for 12 hours or more.

6. The dried precipitate was baked in air for 5 hours at 500°C with a baking rate of 100°C/hour.

7. Thirty three grams of the baked precipitate, 24g of $\text{Mg}(\text{OH})_2$ and 5.7g of graphite carbon were mixed and compression molded at 90kN for 5 minutes to form a molded product with a size of 20mm ϕ .

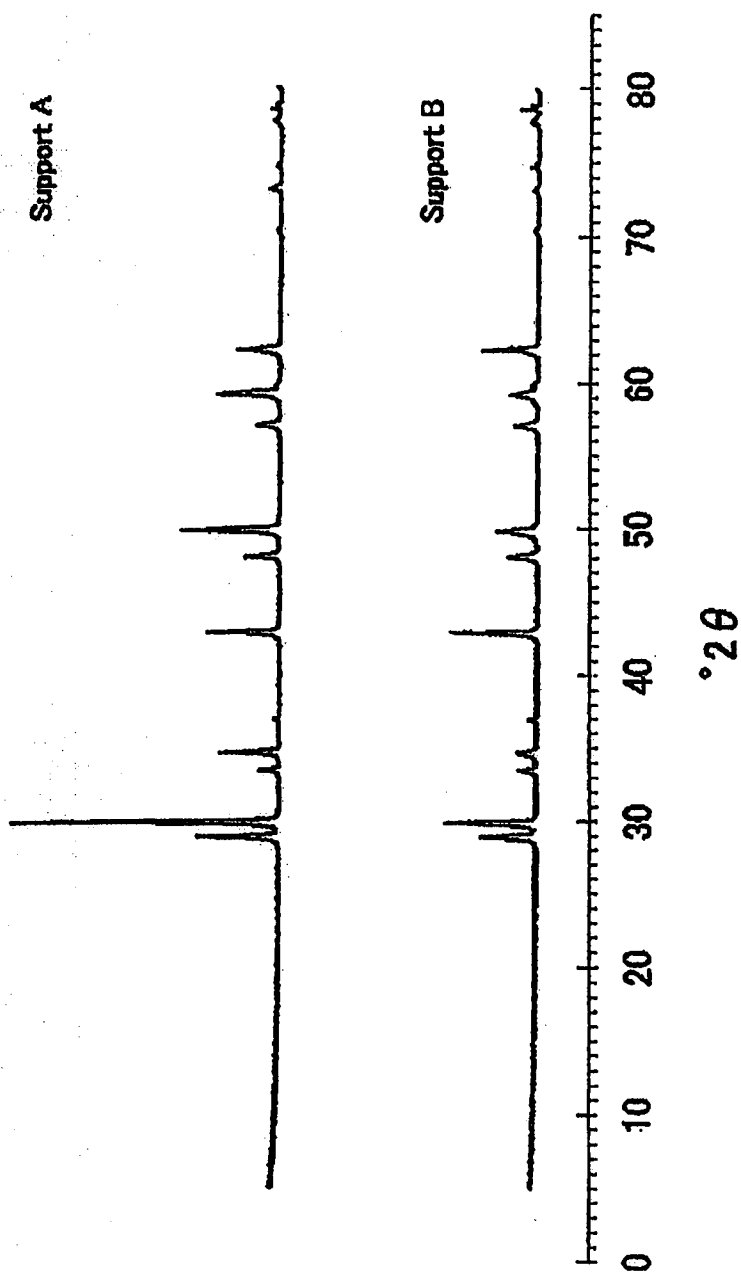
8. The molded product was baked in air for 5 hours at 500°C with a baking rate of 100°C/hour.

9. The baked, molded product (catalyst support B) had a second/first ingredient ratio of 0.213 mol/mol, and a third/first ingredient ratio of 0.170 mol/mol.

X-ray diffraction spectra of catalyst support A and catalyst support B

X-ray diffraction spectra of support A and support B were measured and compared with each other. Good accordance of the spectra will show that support A and support B had substantially the same crystal structure. See the following figure.

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Preparation of catalyst using catalyst support A

1. Support A prepared above was ground and then sieved to have a size of 0.36 to 0.43mmF, which is used as a carrier in the following steps.

2. Onto 4.00g of the prepared carrier was dropped 210mg of an aqueous solution of rhodium acetate with a concentration of 39000ppm to impregnate the carrier with the rhodium acetate solution and the impregnated carrier was dried in air for 1 hour at room temperature.

3. The dried, impregnated carrier was further dried in air for 16 hours at 60°C and subsequently, the further dried, impregnated carrier was baked in air for 3 hours at 950° C with a baking rate of 100°C/hour to obtain the catalyst with a rhodium content of 2000ppm.

Synthesis gas manufacturing test

Synthesis gas manufacturing test was carried out by using the above-obtained catalyst under the same conditions as in Examples 1 to 6 of the present application. The test results are shown below:

Methane conversion rate: 69.9%

CO selectivity: 84.5%

H₂ selectivity: 90.1%

As seen above, the catalyst, which is substantially equivalent to the catalyst of Wu, exhibited a lower performance in terms of methane conversion rate and CO selectivity; but it exhibited a comparable performance in terms of H₂ selectivity.

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6. From the attached experimental results, I have concluded, among other things, that the catalyst of Wu is not adapted for manufacturing synthesis gas with selectivities of at least 90% for both of CO and H₂ particularly for CO, though it may be capable of causing catalytic partial oxidation of methane.

The undersigned declares that all statements made herein of his own knowledge are true, and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under §1001 of Title 18 of the United States Code and that willful false statements may jeopardize the validity of the application or any patent issued thereon.

Takeshi Minami

Takeshi Minami

Signed this 17th day of June, 2009.